

Spin holography II

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Abstract

The interference of the directly emitted photoelectron wave and the wave scattered coherently by neighboring atoms gives holographic fringes in the photoelectron emission intensity $I(\hat{\mathbf{R}})$. In the electron emission holography technique in surface physics, $I(\hat{\mathbf{R}})$ is inverted holographically to give a 3D-image of the environment of the source atom. Earlier [1]–[2], we pointed out that the polarization pattern $\mathbf{P}(\hat{\mathbf{R}})$ similarly can be viewed as a hologram of the spin environment of the source atom by virtue of the exchange scattering of the photoelectron by the neighboring atoms. In this paper, we point out that spin-orbit correlations in the photoelectron initial state are responsible for holographic spin-dependent contributions to the intensity hologram $I(\hat{\mathbf{R}})$, even if the directly emitted photoelectrons are unpolarized. This remarkable result implies that the emission intensity contains spin information just as the polarization pattern $\mathbf{P}(\hat{\mathbf{R}})$. Although the spin dependent signal in the

hologram is rather small ($\sim 5\%$ in most cases of interest), we show how spin information can be extracted from the intensity hologram, making use of the point symmetry of the environment of the source atom. This way of analyzing photoelectron intensity holograms to extract short-range spin information opens up a new avenue for surface magnetism studies.

I. INTRODUCTION

A photoelectron emitted from an atom may be scattered by neighboring atoms. The scattered waves interfere with the unscattered wave and the intensity pattern at large distances, $I(\hat{\mathbf{R}})$, forms a hologram which can be analyzed to yield an image of the neighborhood of the electron emitting atom [3]–[5].

If the neighboring atoms have spins, then exchange scattering results in the polarization of the outgoing electrons and in addition to the intensity $I(\hat{\mathbf{R}})$, the polarization pattern $\mathbf{P}(\hat{\mathbf{R}})$ also forms a hologram which can be analyzed to yield an image of the spins of the neighboring atoms. While this scheme of ‘spin holography’ is elegant, it suffers from the fact that present day electron polarimeters are very inefficient (eff. $\sim 10^{-4}$) detracting from the practical utility of this way of imaging the spins. Therefore, the existence of an alternate method to extract the spin information from the intensity hologram $I(\hat{\mathbf{R}})$, such as the scheme that we discuss in this paper, is of interest.

Spin holography is a holography scheme yielding spin information on the near-neighborhood probed by electron holography. This is in contrast to the long range order and bulk magnetism determinations in magnetic diffraction methods using neutrons or X-rays. Since the effect of exchange interaction on the electron-atom scattering becomes less pronounced as the kinetic energy of the electron increases, it is likely that spin holography will prove to be most useful if the photoelectron has relatively low energy (~ 100 -200 eV). In this energy range, the mean-free path of the electron is of the order of a few nanometers and only atoms within that distance of the photoelectron source atom will contribute significantly to the hologram. Therefore, we expect that electron holographic spin determination will be useful as a probe of surface magnetism, with the source atoms lying within the first few surface layers or as adatoms adhering to the surface. As an example where holographic spin determination is useful, we can use adatom sources on the surface of a ferromagnet to determine the spins of the first few layers, while the surface information is difficult to obtain from ‘diffraction’ measurements.

Consider the example of a substrate with the first two layers magnetized, layer #1 having all its atoms with spin s_a and layer #2 having spin s_b . Then while the holographic method with the sources, e.g. adatoms on the top surface, would yield a proper image of the neighboring atoms on the two layers, diffraction measurements can only distinguish the actual magnetic surface $(1_a, 2_b)$ from the surface with the spins of the layers reversed $(1_b, 2_a)$, by detailed comparison of the diffraction data with numerical multiple scattering calculations.

Clearly, if the photoelectrons are *polarized* then the *intensity* holograms, $I(\hat{\mathbf{R}})$, are sensitive to the spins of the neighboring scatterers. However, a new and important feature of the ‘spherical wave holograms’ is that the photoelectrons need not be polarized for $I(\hat{\mathbf{R}})$ to be used to image the spins of the neighboring atoms: unpolarized electrons suffice if there are spin-orbit correlations. As a consequence, if the photoelectrons are ejected from inner core spin-orbit split subshells (e.g. L_2 or L_3), *even by linearly polarized or unpolarized incident light*, then as we show below, their intensity holograms $I(\hat{\mathbf{R}})$, can be analyzed to image the spins. This is an interesting and important new result. It is important that linearly polarized synchrotron radiation can be used to image the spins. It is interesting for the same reason – one is surprised (at least at first) that linearly polarized light ejecting electrons from a complete inner atomic subshell gives an intensity pattern with a linear dependence on the spins \mathbf{s}_i of the neighboring atoms. (Indeed, the analysis of these matters lead us to interesting new spinor calculus results [7]). Below, we discuss the cases where linearly polarized, circularly polarized and unpolarized light is used to eject the photoelectrons.

The paper is organized as follows. In section II, we calculate the photoelectron hologram, including the exchange scattering contributions which give a linear dependence on the spins of the atoms in the near-neighborhood of the photoelectron source atom. Often the emitting atoms are centers of point symmetry, e.g. C_{nv} , for the crystalline surfaces being investigated. In those cases, we can use symmetry to aid in spin determination, as we discuss in section III. We then proceed in section IV to illustrate the technique for a specific example of a

C_{4v} environment. In section V, we discuss various schemes to extract spin information from the holograms. Finally, in section VI, we comment on the interesting case of rare earth magnetism, for which the theory of spin holography, as formulated below, needs to be generalized. We conclude and summarize the results in section VII.

II. ELECTRON HOLOGRAMS

Initially, to be definite, we consider photoelectrons emitted from a $p_{1/2}$ subshell of a source atom following the absorption of photons linearly polarized in the z-direction. The $p_{1/2}$ subshell consists of two electrons with magnetic quantum numbers $m_j = +1/2$ and $m_j = -1/2$ respectively. The primary electron wave emitted from the m_j -state is a spherically outgoing wave of wave number k ,

$$|\psi_{m_j}^0\rangle \sim |\chi_{m_j}(\hat{\mathbf{r}})\rangle \frac{\exp(ikr)}{r}, \quad (1)$$

where $|\chi_{m_j}(\hat{\mathbf{r}})\rangle$ is the appropriate angular dependent spinor of the primary electron wave. In the reference frame pictured in Fig.(1), these spinors (in the dipole approximation) are proportional to

$$\begin{aligned} |\chi_{1/2}(\hat{\mathbf{R}})\rangle &= \begin{pmatrix} \cos^2(\theta) + c' \\ \sin(\theta) \cos(\theta) \exp(i\phi) \end{pmatrix}; \\ |\chi_{-1/2}(\hat{\mathbf{R}})\rangle &= \begin{pmatrix} \sin(\theta) \cos(\theta) \exp(-i\phi) \\ \cos^2(\theta) + c' \end{pmatrix}, \end{aligned} \quad (2)$$

where θ is the usual polar angle and ϕ the azimuthal angle of $\hat{\mathbf{R}}$ in Eq.(2), and in the following, $c' = (c - 1)/3$, where c is the ratio of the radial matrix elements M_0 (M_2) and corresponding phase shifts δ_0 (δ_2) for emission into the continuum s- or d-states, $c = (M_0/M_2) \exp[i(\delta_0 - \delta_2)]$.

The primary wave is scattered by an atom i , giving a scattered wave $|\psi_{m_j}^i\rangle$. We suppose that $k r_i \gg 1$ and approximate the scattered wave by that of an incident plane wave with

wave vector $k\hat{\mathbf{r}}_i$, see [9]– [11] for a discussion of this approximation in holography. In this case $|\psi_{m_j}^i\rangle$ takes the form,

$$|\psi_{m_j}^i(\hat{\mathbf{R}})\rangle = \frac{\exp(ikr_i)}{r_i} f(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i) \exp(-ik\hat{\mathbf{R}} \cdot \hat{\mathbf{r}}_i) |\chi_{m_j}(\hat{\mathbf{r}}_i)\rangle \frac{\exp(ikR)}{R}, \quad (3)$$

where $f(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i)$ is the scattering amplitude for the scattering of the photoelectron, incident along the $\hat{\mathbf{r}}_i$ -direction and scattered in the $\hat{\mathbf{R}}$ -direction.

If now we neglect multiple scattering, then $|\psi_{m_j}(\mathbf{r})\rangle = |\psi_{m_j}^0(\mathbf{r})\rangle + \sum_i |\psi_{m_j}^i(\mathbf{r})\rangle$ represents the photoelectrons emitted from the m_j -state. The number of electrons emitted in direction $\hat{\mathbf{R}}$, per unit of solid angle is, up to a constant of proportionality which depends on the intensity of the incident photons and the counting time, equal to the ‘intensity’ $I(\hat{\mathbf{R}}) \equiv R^2 \sum_{m_j} \langle \psi_{m_j}(\mathbf{R}) | \psi_{m_j}(\mathbf{R}) \rangle$. Using the above expression we find

$$I(\hat{\mathbf{R}}) = \sum_{m_j} \left[\langle \chi_{m_j}(\hat{\mathbf{R}}) | \chi_{m_j}(\hat{\mathbf{R}}) \rangle + \sum_i 2 \operatorname{Re} ([\exp(ikr_i)/r_i] \langle \chi_{m_j}(\hat{\mathbf{R}}) | f(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i) | \chi_{m_j}(\hat{\mathbf{r}}_i) \rangle \exp(-ik\hat{\mathbf{R}} \cdot \hat{\mathbf{r}}_i)) + \dots \right] \quad (4)$$

In (4) we keep only zero and first order terms in the scattering amplitudes, f , neglecting the effects of multiple scattering and self interference terms (such terms tend to degrade the holographic images and can be partially eliminated by using special kernels in the holographic transform, as well as sums over holograms collected at different energies, as has been discussed extensively in the literature [12]– [14]).

The phase factor $\exp(-ik\hat{\mathbf{R}} \cdot \hat{\mathbf{r}}_i)$ in Eq.(4) gives holographic fringes in the angular intensity pattern; and transforms of $I(\hat{\mathbf{R}})$ by suitable kernels proportional to $\exp(ik\hat{\mathbf{R}} \cdot \mathbf{r})$, where \mathbf{r} is the parameter of the transform, peak in the vicinities of $\mathbf{r} = \mathbf{r}_i$, yielding atomic images.

If scattering atom i has a spin, then the scattering amplitude for coherent scattering, $f(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i)$, depends on the thermally averaged expectation value of the spin vector, \mathbf{s}_i , of the scattering atom,

$$f(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i) = f_0(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i) + f_s(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i) \sigma \cdot \mathbf{s}_i, \quad (5)$$

where the first term is the spin independent contribution f_0 and the second term is the exchange contribution, proportional to the scalar product of \mathbf{s}_i with the Pauli-spin operator

σ of the photoelectron. If we could treat the scattering atoms as spherically symmetric then f_0 and f_s would only depend on $\hat{\mathbf{R}} \cdot \hat{\mathbf{r}}_i$. In reality, the valence shell electron distributions are affected by the presence of the neighboring atoms. The charge (and spin) distribution within a Wigner-Seitz cell representing an atom is not spherically symmetric about the cell's center. This is particularly true for the valence shell electrons having the uncompensated spins which is our primary concern. In the copper oxides of interest in the high T_c materials, for example, the uncompensated spins are believed to occupy $x^2 - y^2$ d-orbitals on the copper ions. Furthermore, we note that the scattering amplitude (5) describes exchange scattering by a magnetic ion if the orbital moments are 'quenched' and the spin direction is free to change without changing the spatial distribution of the charge or spin distributions. This description is accurate for transition elements, but fails for rare earth ions (except for Gd^{3+} , in which case $L = 0$). In that case, there is strong spin-orbit correlation and both f_0 and f_s in Eq.(5) must themselves be taken to be functions of \mathbf{s}_i . In the following, we shall assume a scattering amplitude of the form (5) and we will return to the more complicated but important case of rare earth ions later (section VI). In any case, due to the spin dependence of the scattering amplitude, it should be evaluated inside the spinor bracket of Eq.(4). With the scattering amplitude (5) we obtain

$$I(\hat{\mathbf{R}}) = \langle \langle \hat{\mathbf{R}} | \hat{\mathbf{R}} \rangle \rangle + 2\text{Re} \left(\sum_i [\exp(ikr_i)/r_i] \times \right. \\ \left. [f_0(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i) \langle \langle \hat{\mathbf{R}} | \hat{\mathbf{r}}_i \rangle \rangle + f_s(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i) \langle \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_i \rangle \rangle \cdot \mathbf{s}_i] \exp(-ik\mathbf{R} \cdot \mathbf{r}_i) + \dots \right) , \quad (6)$$

where we represent the sum over the initial electron states m_j of the 'spinor interference brackets' by

$$\langle \langle \hat{\mathbf{R}} | \hat{\mathbf{r}}_i \rangle \rangle = \sum_{m_j} \langle \chi_{m_j}(\hat{\mathbf{R}}) | \chi_{m_j}(\hat{\mathbf{r}}_i) \rangle , \\ \langle \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_i \rangle \rangle = \sum_{m_j} \langle \chi_{m_j}(\hat{\mathbf{R}}) | \sigma | \chi_{m_j}(\hat{\mathbf{r}}_i) \rangle . \quad (7)$$

Eq.(6) includes an interference term that is proportional to f_s and the spin \mathbf{s}_i of the scattering atom so that the intensity is sensitive to the spins of the neighboring atoms if the 'spin interference matrix element', $\langle \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_i \rangle \rangle$, does not vanish. Generally, even for

linearly polarized light (or indeed, unpolarized light) the spin interference matrix element *does not vanish* if the electron is ejected from a spin-orbit split sublevel.

In our example of photoemission from a $p_{1/2}$ level, the interference matrix elements for photons of arbitrary polarization $\hat{\epsilon}$ ($\hat{\epsilon} = \hat{\mathbf{z}}$, for z-linearly polarized, $\hat{\epsilon} = (\hat{\mathbf{x}} \pm i\hat{\mathbf{y}})/\sqrt{2}$ for right- and left-hand polarized light etc) are equal to

$$\begin{aligned} \langle \langle \hat{\mathbf{R}} | \hat{\mathbf{r}} \rangle \rangle &= 2 \{ (\hat{\epsilon}^* \cdot \hat{\mathbf{R}}) (\hat{\epsilon} \cdot \hat{\mathbf{r}}) (\hat{\mathbf{r}} \cdot \hat{\mathbf{R}}) + |c'|^2 + c'^* (\hat{\epsilon} \cdot \hat{\mathbf{r}}) (\hat{\epsilon}^* \cdot \hat{\mathbf{r}}) + c' (\hat{\epsilon}^* \cdot \hat{\mathbf{R}}) (\hat{\epsilon} \cdot \hat{\mathbf{R}}) \} , \\ \langle \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}} \rangle \rangle &= -2 i \{ |c'|^2 (\hat{\epsilon}^* \times \hat{\epsilon}) - c' (\hat{\epsilon} \times \hat{\mathbf{R}}) (\hat{\epsilon}^* \cdot \hat{\mathbf{R}}) + c'^* (\hat{\epsilon}^* \times \hat{\mathbf{r}}) (\hat{\epsilon} \cdot \hat{\mathbf{r}}) \\ &\quad + (\hat{\epsilon}^* \cdot \hat{\mathbf{R}}) (\hat{\epsilon} \cdot \hat{\mathbf{r}}) (\hat{\mathbf{R}} \times \hat{\mathbf{r}}) \} . \end{aligned} \quad (8)$$

For emission from a filled $p_{3/2}$ -shell the intensity interference matrix element, $\langle \langle \hat{\mathbf{R}} | \hat{\mathbf{r}} \rangle \rangle$, is twice the intensity matrix element of Eq.(8), as a consequence of the $p_{3/2}$ -shell having twice as many electrons. Similarly, the $p_{3/2}$ spin interference matrix element, $\langle \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}} \rangle \rangle$ is the negative of the spin interference matrix element shown on the last line of Eq.(8) [15], which follows from the fact that the $p_{3/2}$ and $p_{1/2}$ spin matrix elements add up to the spin matrix element for photoemission from a p-shell which, in the absence of spin-orbit interaction, vanishes.

It is a feature of spherical wave electron holography that the interference between electron waves emitted in *different* directions is recorded. As a consequence, it is not the spin polarization of the electrons emitted in the direction of the scattering atom $\hat{\mathbf{r}}_i$ that gives the holographic spin dependence, but the interference matrix element of the emission directions $\hat{\mathbf{R}}$ and $\hat{\mathbf{r}}_i$, $\langle \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_i \rangle \rangle$. The occurrence of these quantities in our theory leads to interesting new aspects of angular momentum theory.

That the spin matrix element is of the form of Eq.(8) can be understood from general considerations – the matrix element must be ‘bilinear’ in the photon polarization, meaning that each term contains $\hat{\epsilon}$ and $\hat{\epsilon}^*$. Furthermore, the spin matrix element must be a pseudo vector made up of $\hat{\epsilon}$, $\hat{\epsilon}^*$, $\hat{\mathbf{R}}$, and $\hat{\mathbf{r}}_i$. In the dipole approximation, the wave emitted from a p-shell is an admixture of an s and a d-wave, from which follows that each term contains $\hat{\mathbf{R}}$ and $\hat{\mathbf{r}}_i$ twice or not at all. Based on these considerations we can deduce the general form of

the individual terms that make up the $p_{1/2}$ (or $p_{3/2}$) spin matrix element.

Some remarkable results follow from Eq.(8). For example, the spin interference matrix element for z-linearly polarized light does not vanish :

$$\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{r}}_i\rangle\rangle_{\mathbf{z}} = -2 i \{ -c'(\hat{\mathbf{z}} \times \hat{\mathbf{R}}) (\hat{\mathbf{z}} \cdot \hat{\mathbf{R}}) + c'^*(\hat{\mathbf{z}} \times \hat{\mathbf{r}}_i) (\hat{\mathbf{z}} \cdot \hat{\mathbf{r}}_i) + (\hat{\mathbf{z}} \cdot \hat{\mathbf{R}}) (\hat{\mathbf{z}} \cdot \hat{\mathbf{r}}_i) (\hat{\mathbf{R}} \times \hat{\mathbf{r}}_i) \} , \quad (9)$$

implying that linearly polarized photons give spin dependent photoelectron intensities. The expected spin of the directly emitted photoelectrons in the \mathbf{R} -direction is proportional to $\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{R}}\rangle\rangle$, which can be obtained from Eq.(9), putting $\hat{\mathbf{r}}_i \rightarrow \hat{\mathbf{R}}$. This gives an electron spin polarization that is proportional to

$$\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{R}}\rangle\rangle_{\mathbf{z}} = -\frac{4}{3} \sin(\delta_0 - \delta_2) \frac{M_0}{M_2} (\hat{\mathbf{z}} \times \hat{\mathbf{R}}) (\hat{\mathbf{z}} \cdot \hat{\mathbf{R}}) , \quad (10)$$

where we replaced the imaginary part of the c' -parameter by its expression in terms of the phase shifts and the radial dipole matrix elements. The result of Eq.(10) makes it clear that the resulting spin polarization is in fact caused by the interference of the outgoing s and d-waves, as was pointed out previously [16]. Nevertheless, even if the spin polarization (10) of the unscattered photoelectrons vanish, because, for example, the electron is emitted as a pure d-wave, the interference spin matrix element does *not* vanish. The pure d-wave photoemission corresponds to $c' = -1/3$ giving

$$\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{r}}_i\rangle\rangle_{\mathbf{z}} = -2 i \left\{ \frac{1}{3}(\hat{\mathbf{z}} \times \hat{\mathbf{R}}) (\hat{\mathbf{z}} \cdot \hat{\mathbf{R}}) - \frac{1}{3}(\hat{\mathbf{z}} \times \hat{\mathbf{r}}_i) (\hat{\mathbf{z}} \cdot \hat{\mathbf{r}}_i) + (\hat{\mathbf{z}} \cdot \hat{\mathbf{R}}) (\hat{\mathbf{z}} \cdot \hat{\mathbf{r}}_i) (\hat{\mathbf{R}} \times \hat{\mathbf{r}}_i) \right\} . \quad (11)$$

Thus the d-wave photoelectron incident upon and scattered from the scattering atom is not spin polarized, (i.e. $\langle\langle\hat{\mathbf{r}}_i|\sigma|\hat{\mathbf{r}}_i\rangle\rangle = 0$), nor is the unscattered wave emitted in the $\hat{\mathbf{R}}$ -direction polarized, (i.e. $\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{R}}\rangle\rangle = 0$); but the interference between the photoelectron waves emitted in the \mathbf{r}_i -direction and scattered into the direction $\hat{\mathbf{R}}$ with the waves emitted directly in that direction does result in a polarization and gives a photoelectron intensity sensitive to the spin of the scattering atom. Another interesting consequence of this angular correlation in the photoelectron spin is that the interference of the directly emitted and

scattered electron waves gives a finite spin polarization to the photoelectrons, even though the directly emitted electrons are spin unpolarized and the scattering atom is not magnetic.

Perhaps even more surprising than the fact that incident linearly polarized light gives rise to spin-sensitive holograms is that so does unpolarized light. Note that unpolarized light incident along the z-direction, say, on the source atom gives photoelectron intensities equal to the sum of those for x- and y-linearly polarized beams. Now the sum of x-,y- and z-polarized beams gives

$$\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{r}}_i\rangle\rangle = -2i(\hat{\mathbf{R}}\times\hat{\mathbf{r}}_i)(\hat{\mathbf{r}}_i\cdot\hat{\mathbf{R}}) , \quad (12)$$

thus,

$$\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{r}}_i\rangle\rangle_{\mathbf{x}} + \langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{r}}_i\rangle\rangle_{\mathbf{y}} = -\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{r}}_i\rangle\rangle_{\mathbf{z}} - 2i(\hat{\mathbf{R}}\times\hat{\mathbf{r}}_i)(\hat{\mathbf{r}}_i\cdot\hat{\mathbf{R}}) , \quad (13)$$

and substituting from Eq.(9) we see that unpolarized light also gives a non-vanishing spin contribution to the photoelectron intensity and thus can serve to photoemit electrons with an intensity hologram that is sensitive to the spins of the nearby atoms.

These surprising results are caused by the spin-orbit correlation in the initial shell, which is preserved in the emitted d-waves. The remarkable consequences clearly indicate that the interference matrix elements contain novel and interesting physics.

Returning to the intensity hologram of Eq.(6), it is clear from the subsequent discussion of the interference matrix elements that the spin contributions to the intensity hologram don't vanish if the electrons are emitted from an atomic spin-orbit split shell. The spin contributions are, however, relatively small, only making an addition of the order of $|f_s\mathbf{s}_i/f_0|$ ($\simeq 0.05$, e.g., for electron scattering by an iron atom at 100 eV) to the ordinary, spin independent 'charge' hologram. *The challenge then is to extract the spin information from the hologram.*

III. SPIN ANALYSIS

The question becomes how to isolate the spin dependent part of the hologram from the much larger ‘charge’ part background.

Referring back to Eq.(6), if we were able to reverse all the \mathbf{s}_i ’s of the sample then the difference hologram would contain only the spin dependent terms. Reversing the sample’s spins, however, would be feasible only for ferro- or ferrimagnetic samples. Alternatively, the difference of holograms below and above the magnetic ordering temperatures can serve to determine the spins in the cases where we can assume that the change of the spin independent factors in Eq.(6) is small.

Other than those two methods depending on changing the magnetization of the sample by changing the magnetic field or the temperature, which may not be feasible, there are other methods which can serve our purposes:

If the primary wave, (2), had only an s-wave component rather than the d-s mixture, then only the $|c'|^2$ terms in Eq.(8) would be present. In that case $\langle \hat{\mathbf{R}} | \hat{\mathbf{r}}_i \rangle \sim |c'|^2$, $\langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_i \rangle_{\pm} \sim \pm |c'|^2 \hat{\mathbf{z}}$ for $\hat{\mathbf{e}} = (\hat{\mathbf{x}} \pm \hat{\mathbf{y}})/\sqrt{2}$, and the difference holograms for incident right and left-hand circularly polarized light around, successively x,y and z-axes would determine all three components of the spin vectors. In fact, however, the s-wave component is almost an order of magnitude smaller than the dominant d-wave contribution in the case of photoemission from a p-shell at the relevant emission energies ($\sim 100 - 200$ eV). Furthermore, we note that the extraction of the spin terms from right-left polarization differences contain spin independent ‘charge’ contributions. Indeed, with Eq.(8) we find

$$\langle \hat{\mathbf{R}} | \hat{\mathbf{r}}_i \rangle_+ - \langle \hat{\mathbf{R}} | \hat{\mathbf{r}}_i \rangle_- = -2i \left[(\hat{\mathbf{r}}_i \cdot \hat{\mathbf{R}}) (\hat{\mathbf{R}} \times \hat{\mathbf{r}}_i) \cdot \hat{\mathbf{z}} \right] , \quad (14)$$

and referring back to Eq.(6), we see that the difference hologram does indeed contain ‘charge’ contributions.

We can, however, use *symmetry* to aid in spin determination. Often the emitting atoms are centers of point symmetry, e.g. C_{nv} , for the crystalline surfaces being investigated. Any

differences detected after subjecting the hologram to a point symmetry group operation is then the result of the symmetry breaking spin contributions. Based on this general idea, we work out practical schemes to construct ‘spin holograms’. For the sake of simplicity, we start by illustrating the main idea for the case of reflection symmetry in subsection III A. In subsection III B, we approach the problem from a more general perspective.

A. Reflection Spin Holography

Let $I(\hat{\mathbf{R}}, \hat{\epsilon}; m)$ represent the photoelectron intensity emitted in the $\hat{\mathbf{R}}$ -direction, for photon polarization $\hat{\epsilon}$, matter state m , and let $I(\hat{\mathbf{R}}', \hat{\epsilon}'; m')$ be the corresponding quantities following a spatial reflection. Since the Hamiltonian is invariant under reflections,

$$I(\hat{\mathbf{R}}, \hat{\epsilon}; m) = I(\hat{\mathbf{R}}', \hat{\epsilon}'; m') . \quad (15)$$

Also,

$$I(\hat{\mathbf{R}}', \hat{\epsilon}'; m) = I(\hat{\mathbf{R}}, \hat{\epsilon}; m') , \quad (16)$$

since the square of the reflection is the unit operator. Now ‘ m ’ represents the initial state of the source atom, centered at the origin, and the various neighboring atoms centered at \mathbf{r}_i , with spins \mathbf{s}_i , and other properties, including valence electron spatial distributions, o_i : $m = [\mathbf{r}_1, \mathbf{s}_1, o_1; \mathbf{r}_2, \mathbf{s}_2, o_2, \dots, \mathbf{r}_i, \mathbf{s}_i, o_i; \dots]$, $m' = [\mathbf{r}'_1, \mathbf{s}'_1, o'_1; \dots]$. Since \mathbf{s} is an axial vector, $\mathbf{s}'_{//} = -\mathbf{s}_{//}$, $\mathbf{s}'_{\perp} = \mathbf{s}_{\perp}$, where the $//$ and \perp subscripts refer to components respectively parallel and perpendicular to the plane of reflection.

If the source atom is in a site with C_{nv} point symmetry, magnetic ordering of the neighboring atoms may break that symmetry, and the difference hologram,

$$\begin{aligned} I_V(\hat{\mathbf{R}}, \hat{\epsilon}; m) &\equiv \frac{1}{2} [I(\hat{\mathbf{R}}, \hat{\epsilon}; m) - I(\hat{\mathbf{R}}', \hat{\epsilon}'; m)] , \\ &= \frac{1}{2} [I(\hat{\mathbf{R}}, \hat{\epsilon}; m) - I(\mathbf{R}, \hat{\epsilon}; m')] , \end{aligned} \quad (17)$$

contains only the symmetry breaking spin terms if the reflection plane, V , is a symmetry plane. This is so because if atom ‘ j ’ is carried into atom ‘ k ’ by the reflection then $o'_j = o_k$,

by symmetry, and the non-spin dependent part of the hologram, the ‘charge’ hologram is unchanged by the reflection.

If atom ‘i’ lies in the reflection plane then $\mathbf{r}_i = \mathbf{r}'_i$, $o_i = o'_i$, $\mathbf{s}_{i, //} = -\mathbf{s}'_{i, //}$, $\mathbf{s}_{i, \perp} = \mathbf{s}'_{i, \perp}$, and the contribution of ‘i’ to the difference hologram I_V is (see Eq.(6))

$$I_V(\hat{\mathbf{R}}) = 2 \operatorname{Re}\{[\exp(ikr_i)/r_i] f_s(\mathbf{R}, \mathbf{r}_i) \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_i \rangle \cdot \mathbf{s}_{i, //} \exp(-ik\hat{\mathbf{R}} \cdot \mathbf{r}_i)\} . \quad (18)$$

On the other hand, if atom ‘j’ does not lie in the reflection plane ‘V’ and is carried into atom ‘k’ by the reflection then $\mathbf{s}_k \rightarrow \frac{1}{2}(\mathbf{s}_k - \mathbf{s}'_j) = \frac{1}{2}(\mathbf{s}_k + \mathbf{s}_{j, //} - \mathbf{s}_{j, \perp})$, and the contribution of site ‘k’ to I_V becomes

$$I_V(\hat{\mathbf{R}}) = 2 \operatorname{Re}\{[\exp(ikr_k)/r_k] f_s(\mathbf{R}, \mathbf{r}_k) \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_k \rangle \cdot \frac{1}{2}(\mathbf{s}_k + \mathbf{s}_{j, //} - \mathbf{s}_{j, \perp}) \exp(-ik\hat{\mathbf{R}} \cdot \mathbf{r}_k)\} . \quad (19)$$

The hologram I_V given by summing the in-plane, Eq.(18), and out of plane, Eq.(19), terms can then be transformed, as we shall discuss later, to determine components of the spins of the various atoms.

B. Projection Spin Holography

The above considerations regarding reflections illustrate the use of symmetry to obtain spin holograms. If G is an element, a reflection or a rotation, of a point symmetry group which leaves the charge or chemical neighborhood invariant, then the difference hologram involving G has only the symmetry breaking spin contributions. The argument is essentially that given above. For example, the difference between the emission intensity $I(\hat{\mathbf{R}}, \hat{\epsilon}; m)$ measured in direction $\hat{\mathbf{R}}$ with incident photons of polarization $\hat{\epsilon}$ and the intensity measured in direction $G[\hat{\mathbf{R}}]$ with photons of polarization $G[\hat{\epsilon}]$, $I(G[\hat{\mathbf{R}}], G[\hat{\epsilon}]; m)$ contains only spin contributions because the charge contributions cancel.

To see that, we note that the Hamiltonian is invariant under all of the transformations of the euclidean group (except for the small parity non-conserving terms which are irrelevant for our considerations). Thus, it follows that

$$I(\hat{\mathbf{R}}, \hat{\epsilon}; m) = I(G[\hat{\mathbf{R}}], G[\hat{\epsilon}]; G[m]) , \quad (20)$$

which is the generalization of (15), and where, as before, m represents the initial state of the source atom and the neighboring atoms, $m = [\mathbf{r}_1, \mathbf{s}_1, o_1; \mathbf{r}_2, \mathbf{s}_2; o_2, \dots]$. Also, as in Eq.(16),

$$I(G^{-1}[\hat{\mathbf{R}}], G^{-1}[\hat{\epsilon}]; m) = I(\hat{\mathbf{R}}, \hat{\epsilon}; G[m]) , \quad (21)$$

where $G[m] = [G[\mathbf{r}_1], G[\mathbf{s}_1], G[o_1]; \dots, G[\mathbf{r}_j], G[\mathbf{s}_j], G[o_j]; \dots]$. Since G and G^{-1} belong to the symmetry group, for every atom j there is an atom k for which $\mathbf{r}_j = G[\mathbf{r}_k]$ (and $o_j = G[o_k]$). If we denote the label k by $k = g[j]$, then $G[\mathbf{s}_k] = G[\mathbf{s}_{g[j]}]$ and we can represent the transformed initial state as $G[m] = [\mathbf{r}_1, G[\mathbf{s}_{g[1]}], o_1; \dots; \mathbf{r}_j, G[\mathbf{s}_{g[j]}], o_j; \dots]$, representing a neighborhood which has the same ‘charge’ distribution as m , but a different spin arrangement: \mathbf{s}_1 appears now as $G[\mathbf{s}_1]$ at the site of $G[\mathbf{r}_1]$ etc... . Consequently, only spin dependent terms do not cancel in the difference hologram

$$I_G \equiv \frac{1}{2} \left[I(\hat{\mathbf{R}}, \hat{\epsilon}; m) - I(G^{-1}[\hat{\mathbf{R}}], G^{-1}[\hat{\epsilon}]; m) \right] . \quad (22)$$

More generally, any linear combination of the type $\sum_G a(G) I(G^{-1}[\hat{\mathbf{R}}], G^{-1}[\hat{\epsilon}]; m)$, where the sum \sum_G extends over all elements G of the symmetry group of the ‘charge’ environment and where the sum of the coefficients $a(G)$ vanishes, $\sum_G a(G) = 0$, results in a hologram of which the charge contributions cancel out but not the spin terms. In fact, the resulting angular pattern, $\sum_G a(G) I(G[\hat{\mathbf{R}}], G[\hat{\epsilon}]; m)$ is a hologram of the spin arrangement obtained from the actual spin distribution, by replacing the spin \mathbf{s}_j at site \mathbf{r}_j by $\sum_G a(G) G[\mathbf{s}_{g(j)}]$:

$$\begin{aligned} I_a(\hat{\mathbf{R}}, \hat{\epsilon}; m) &\equiv \sum_G a(G) I(G^{-1}[\hat{\mathbf{R}}], G^{-1}[\hat{\epsilon}]; m) = \sum_G a(G) I(\hat{\mathbf{R}}, \hat{\epsilon}; G[m]) \\ &= 2 \sum_j \text{Re}\{[\exp(ikr_j)/r_j] f_s(\hat{\mathbf{R}}, \hat{\mathbf{r}}_j) \exp(-ik\hat{\mathbf{R}} \cdot \hat{\mathbf{r}}_j) \\ &\quad \langle \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_j \rangle \rangle \cdot \sum_G a(G) G[\mathbf{s}_{g[j]}] \} + \dots . \end{aligned} \quad (23)$$

Consider then the ‘star’ of an atom at \mathbf{r}_1 , i.e., the atoms at $(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)$ into which \mathbf{r}_1 is carried by the operations of the group. (Thus for the group C_{nv} , $N=n$, if \mathbf{r}_1 lies in a reflection plane, and $N=2n$, the order of the group, if it doesn’t.) Then the spins on

the N atoms constitute a $3N$ dimensional linear vector space which transforms into itself under the group and which can be decomposed into irreducible subspaces forming bases of the irreducible representation of the group by the well known methods of group theory [17]. We may represent a vector in this $3N$ dimensional space as $\mathbf{s}_{(N)} \equiv \{\mathbf{s}_1, \mathbf{s}_2, \dots; \mathbf{s}_N\} = (\mathbf{s}_{1x}, \mathbf{s}_{1y}, \mathbf{s}_{1z}; \mathbf{s}_{2x}, \mathbf{s}_{2y}, \mathbf{s}_{2z}; \dots)$. From the general principles of group theory, we know that the $\mathbf{s}_{(N)}$ -vector can be decomposed into components that transform according to the irreducible representation ' α ' of the symmetry group. The decomposition, $\mathbf{s}_{(N)} = \sum_{i,\alpha} \mathbf{s}_{(N)}^{(\alpha,i)}$, is achieved by means of the idempotent operator, $e_{ii}^\alpha = \frac{l_\alpha}{h} \sum_G D_{ii}^\alpha(G^{-1})G$, which, when applied to a linear vector space, selects out the (α, i) -component of the vector :

$$\mathbf{s}_{(N)}^{(\alpha,i)} = \left(\frac{l_\alpha}{h} \right) \sum_G D_{ii}^\alpha(G^{-1}) G[\mathbf{s}_{(N)}] , \quad (24)$$

where $D_{ij}^\alpha(G)$ is the matrix representation of G in the irreducible representation α , h is the order of the symmetry group and i takes on values from 1 to l_α , the dimension of the representation.

Returning to the spin hologram of Eq.(23), it is then clear that identifying the $a(G)$ coefficients with $\left(\frac{l_\alpha}{h} \right) D_{ii}^\alpha(G^{-1})$, gives a pattern that is the hologram of the total spin $\mathbf{s}_{(N)}$ projected onto the irreducible (α, i) -mode $\mathbf{s}_{(N)}^{(\alpha,i)}$.

This procedure, projecting out the irreducible modes of the photoelectron hologram,

$$I^{(\alpha,i)}(\hat{\mathbf{R}}, \hat{\epsilon}; m) = \left(\frac{l_\alpha}{h} \right) \sum_G D_{ii}^\alpha(G^{-1}) I(G^{-1}[\hat{\mathbf{R}}], G^{-1}[\hat{\epsilon}]; m) , \quad (25)$$

giving a hologram of the irreducible (α, i) -mode of the spin pattern, is completely general and gives the maximum information that can be obtained from symmetry.

Now we take $\alpha = 1$ for the identical representation, $D^1(G) = 1$, so that the vector $\mathbf{s}_{(N)}^1$,

$$\mathbf{s}_{(N)}^1 = \left(\frac{1}{l} \right) \sum_G G^{-1}[\mathbf{s}_{(N)}] , \quad (26)$$

is invariant under all G 's and thus does not appear in the difference holograms as in (23). Such a vector is invariant under all symmetry operations G and cannot be distinguished from the charge environment using symmetry alone. The number of such linearly independent

invariant vectors is given as usual by the compound character averaged over all the group elements. For C_{nv} there is one such vector for the $N=n$ atom star: $\mathbf{s}_i = C\hat{\mathbf{z}} \times \mathbf{r}_i$. Consequently, out of the $3n$ linearly independent vectors $3n - 1$ can be determined from the holograms (25). For the $N=2n$ star there are three such vectors : in addition to that for the $N=n$ case, there are also $\pm\hat{\mathbf{z}}$, and $\pm\hat{\mathbf{r}}_i$, where the signs for the two semi-stars, each consisting of n atoms carried into each other by the C_n subgroup, differ. In this case, one determines $6n - 3$ linearly independent vectors by our method.

IV. AN EXAMPLE WITH C_{4v} SYMMETRY

We illustrate the projection scheme for photoemission from an atom placed in the environment pictured in Fig.(2). Four identical neighboring atoms are located in the same horizontal plane with azimuthal angles $\phi = 0, \pi/2, \pi$ and $3\pi/2$, placed at equal distance from the emitting atom. The result is a ‘charge’ environment of C_{4v} -symmetry.

The C_{4v} -symmetry group has 8 elements ($h=8$): the identity operation E , 2 rotations in the horizontal plane by $\pi/2$: $\phi \rightarrow \phi + \pi/2$ (C_4) and $\phi \rightarrow \phi - \pi/2$ (C_4'), one rotation by π (C_2), and 4 reflections with respect to the vertical planes of azimuthal angle $\phi = 0$ (s_V'), $\phi = \pi/4$ (s_d), $\phi = \pi/2$ (s_V), $\phi = 3\pi/2$ (s_d'). There are 5 classes $\{E, 2C_4, C_2, 2s_V, 2s_d\}$ and 5 irreducible representations costumarily denoted by $\{A_1, A_2, B_1, B_2, E\}$. For the reader’s convenience, we show the character table of C_{4v} in table I.

The spin arrangement consists of four spin vectors (12 components) which generate a 12-dimensional representation of C_{4v} : $D^{(12)}$. Reducing the 12 components to modes that transform according to the irreducible representations, we find one A_1 , one B_1 , two A_2 , two B_2 and three E -modes:

$$D^{(12)} = 1 A_1 \oplus 1 B_1 \oplus 2 A_2 \oplus 2 B_2 \oplus 3 E . \quad (27)$$

Each spin environment is a linear combination of these modes, pictured in Fig(3). Of the irreducible modes, only the A_1 -mode, invariant under all C_{4v} -operations cannot be determined by means of the projection scheme, the other 11 components are determined. Notice

that one of the A_2 -modes represents a ferromagnetic spin arrangement, with the spins in the z-direction. The in-plane (in the x and y-direction) ferromagnetic spin arrangements are E-modes.

In constructing the spin holograms (25), we take linear combinations of transformed emission patterns, $I(G^{-1}[\hat{\mathbf{R}}], G^{-1}[\hat{\epsilon}]; m)$. If $\epsilon = \epsilon_0 (= \hat{\mathbf{z}})$, then since $G[\hat{\epsilon}_0] = \hat{\epsilon}_0$ for all elements G of C_{4v} , only a single photoelectron hologram, $I_0(\hat{\mathbf{R}})$, must be measured. For notational convenience we indicate in what follows the photon polarization by means of a subscript. With circularly polarized light incident along the z-direction, $\hat{\epsilon} = \hat{\epsilon}_+$, $\hat{\epsilon}_-$, the two intensity holograms, I_+ and I_- , suffice to construct the projection holograms (25).

Representing the symmetry operation by the corresponding change in the azimuthal angle dependence {e.g. $I_0(s_V[\hat{\mathbf{R}}])$ is represented by $I_0(\pi - \phi)$ etc...}, we can write the spin holograms for the A_2 , B_1 and B_2 -modes as

$$\begin{aligned}
I^{A_2}(\hat{\epsilon}_0; \hat{\mathbf{R}}) &= \frac{1}{8} \{I_0(\phi) + I_0(\phi + \pi/2) + I_0(\phi - \pi/2) + I_0(\phi + \pi) \\
&\quad - I_0(\pi - \phi) - I_0(-\phi) - I_0(\pi/2 - \phi) - I_0(-\phi - \pi/2)\} , \\
I^{B_1}(\hat{\epsilon}_0; \hat{\mathbf{R}}) &= \frac{1}{8} \{I_0(\phi) - I_0(\phi + \pi/2) - I_0(\phi - \pi/2) + I_0(\phi + \pi) \\
&\quad + I_0(\pi - \phi) + I_0(-\phi) - I_0(\pi/2 - \phi) - I_0(-\phi - \pi/2)\} , \\
I^{B_2}(\hat{\epsilon}_0; \hat{\mathbf{R}}) &= \frac{1}{8} \{I_0(\phi) - I_0(\phi + \pi/2) - I_0(\phi - \pi/2) + I_0(\phi + \pi) \\
&\quad - I_0(\pi - \phi) - I_0(-\phi) + I_0(\pi/2 - \phi) + I_0(-\phi - \pi/2)\} , \tag{28}
\end{aligned}$$

where we suppressed the dependence on the polar angle, θ , which is invariant under C_{nv} symmetry elements. For the one-dimensional A and B -modes, the coefficients can simply be read from the character table (table I). For the two-dimensional E -representation, on the other hand, we need to construct an actual representation. The result is:

$$\begin{aligned}
I^{(E,1)}(\hat{\epsilon}; \hat{\mathbf{R}}) &= \frac{1}{4} \{I_0(\phi) - I_0(\phi + \pi) + I_0(\pi - \phi) - I_0(-\phi)\} , \\
I^{(E,2)}(\hat{\epsilon}; \hat{\mathbf{R}}) &= \frac{1}{4} \{I_0(\phi) - I_0(\phi + \pi) - I_0(\pi - \phi) + I_0(-\phi)\} . \tag{29}
\end{aligned}$$

The analogous spin holograms for the circularly polarized photons, are

$$\begin{aligned}
I^{A_2}(\hat{\epsilon}_+; \hat{\mathbf{R}}) &= \frac{1}{8} \{ I_+(\phi) + I_+(\phi + \pi/2) + I_+(\phi - \pi/2) + I_+(\phi + \pi) \\
&\quad - I_-(\pi - \phi) - I_-(-\phi) - I_-(\pi/2 - \phi) - I_-(-\phi - \pi/2) \} , \\
I^{B_1}(\hat{\epsilon}_+; \hat{\mathbf{R}}) &= \frac{1}{8} \{ I_+(\phi) - I_+(\phi + \pi/2) - I_+(\phi - \pi/2) + I_+(\phi + \pi) \\
&\quad + I_-(\pi - \phi) + I_-(-\phi) - I_-(\pi/2 - \phi) - I_-(-\phi - \pi/2) \} , \\
I^{B_2}(\hat{\epsilon}_+; \hat{\mathbf{R}}) &= \frac{1}{8} \{ I_+(\phi) - I_+(\phi + \pi/2) - I_+(\phi - \pi/2) + I_+(\phi + \pi) \\
&\quad - I_-(\pi - \phi) - I_-(-\phi) + I_-(\pi/2 - \phi) + I_-(-\phi - \pi/2) \} . \\
I^{(E,1)}(\hat{\epsilon}_+; \hat{\mathbf{R}}) &= \frac{1}{4} \{ I_+(\phi) - I_+(\phi + \pi) + I_-(\pi - \phi) - I_-(-\phi) \} , \\
I^{(E,2)}(\hat{\epsilon}_+; \hat{\mathbf{R}}) &= \frac{1}{4} \{ I_+(\phi) - I_+(\phi + \pi) - I_-(\pi - \phi) + I_-(-\phi) \} . \tag{30}
\end{aligned}$$

Eqs. (28), (29) and (30) constitute an analysis of the measured hologram $I(\hat{\mathbf{R}}, \hat{\epsilon}, m)$ into its six components (adding the A_1 component to those given) appropriate to C_{4v} -symmetry. The advantage of this mode of analysis is that each component is the hologram of a relatively simple spin configuration $\mathbf{s}_{(N)}^{(\alpha,i)}$, as given in Fig.(3) for each neighboring star of atoms. Determination of the components $\mathbf{s}_{(N)}^{(\alpha,i)}$ then gives $\mathbf{s}_N (= \sum_{(\alpha,i)} \mathbf{s}_{(N)}^{(\alpha,i)})$.

V. EXTRACTING INFORMATION FROM THE SPIN HOLOGRAMS

How then, do we get the $\mathbf{s}_{(N)}^{(\alpha,i)}$ from the hologram $I^{(\alpha,j)}$? There are two main methods. First we can transform the hologram to give images of a sort of the individual atoms. These ‘images’ take the form shown in Eq.(37) below of standing spherical waves centered on the various atoms with strengths proportional to $\mathbf{s}_{(N)}^{(\alpha,i)}$. The advantage of this approach is that it gives a 3D ‘picture’ of the spin distribution on the neighboring atoms. In fact, although these holographic images are of interest in particular for obtaining a first qualitative determination of the spins, we think that the most efficient and accurate spin determination will involve iterative least square fitting of the direct hologram $I^{(\alpha,j)}(\hat{\mathbf{R}})$.

We first discuss the nature of the holographic image. The field of the holographic image of a traditional (optical) spherical wave hologram $I(\hat{\mathbf{R}})$, formed by irradiating the negative of the hologram with a spherical ingoing wave, can be shown [3] to be proportional to

$$\mathcal{I}(\mathbf{r}) = \int I(\hat{\mathbf{R}}) \exp(ik\hat{\mathbf{R}} \cdot \mathbf{r}) d\Omega_{\hat{\mathbf{R}}} \quad , \quad (31)$$

where $d\Omega_{\hat{\mathbf{R}}}$ denotes an infinitesimal solid angle of the emission direction $\hat{\mathbf{R}}$. In electron emission holography, transforms of the type of Eq.(31) are called images, and as we shall see the atoms contributing to the hologram appear in the image as centers of spherical waves.

Consider the simple example of a hologram formed by a spherically symmetric primary wave, $\exp(ikr)/r$, scattered coherently with scattering amplitude f by atoms i at positions \mathbf{r}_i . In the region $k|\mathbf{r} - \mathbf{r}_i| \gg 1$, the wave scattered by atom i is

$$[\exp(ikr_i)/r_i] f(\widehat{\mathbf{r} - \mathbf{r}_i}; \mathbf{r}_i) \exp(ik|\mathbf{r} - \mathbf{r}_i|)/|\mathbf{r} - \mathbf{r}_i| \quad , \quad (32)$$

where $\widehat{\mathbf{r} - \mathbf{r}_i}$ denotes $(\mathbf{r} - \mathbf{r}_i)/|\mathbf{r} - \mathbf{r}_i|$. The interference between the primary and scattered waves then gives holographic fringes in the emission intensity in the far-region, corresponding to

$$\sum_i 2 \operatorname{Re}\{[\exp(ikr_i)/r_i] f(\hat{\mathbf{R}}; \hat{\mathbf{r}}_i) \exp(-ik\hat{\mathbf{R}} \cdot \mathbf{r}_i) \} \quad . \quad (33)$$

Although the exchange scattering and the angular correlations expressed by the interference matrix elements complicates the expressions, we can still recognize Eq.(33) in the projection hologram $I^{(\alpha,j)}$ of Eq.(23), replacing $f(\hat{\mathbf{R}}; \hat{\mathbf{r}}_i)$ by an ‘effective’ scattering amplitude: $f \rightarrow f^{(eff)}$, where

$$f^{(eff)}(\hat{\mathbf{R}}; \hat{\mathbf{r}}_i) = \langle \langle \hat{\mathbf{R}} | \sigma | \hat{\mathbf{r}}_i \rangle \rangle \cdot \mathbf{s}_i^{(\alpha,j)} f_s(\hat{\mathbf{R}}; \hat{\mathbf{r}}_i) \quad , \quad (34)$$

and where $\mathbf{s}_i^{(\alpha,j)}$ is the spin on atom i in the (α, j) -projection of the $\mathbf{s}_{(N)}$ -spin vector. The interference terms (33) then contribute ‘imaging’ terms $\sum_i \mathcal{I}_i(\mathbf{r})$ to the transform, where

$$\mathcal{I}_i(\mathbf{r}) = [\exp(ikr_i)/r_i] \int f^{(eff)}(\hat{\mathbf{R}}; \hat{\mathbf{r}}_i) \exp(ik\hat{\mathbf{R}} \cdot [\mathbf{r} - \mathbf{r}_i]) d\Omega_{\hat{\mathbf{R}}} \quad . \quad (35)$$

We calculate the image by expanding the exponential factor in partial waves, $\exp(ik\hat{\mathbf{R}} \cdot [\mathbf{r} - \mathbf{r}_i]) = 4\pi \sum_{l,m} (i)^l j_l(k|\mathbf{r} - \mathbf{r}_i|) Y_{lm}(\widehat{\mathbf{r} - \mathbf{r}_i}) Y_{lm}^*(\hat{\mathbf{R}})$, where j_l denotes the spherical Bessel functions of the second kind and the Y_{lm} -functions are the spherical harmonics. Similarly,

$f^{(eff)}(\hat{\mathbf{R}}; \hat{\mathbf{r}}_i) = \sum_{LM} F_{LM} Y_{LM}(\hat{\mathbf{R}})$, and assuming for the sake of the argument that the hologram is collected over the full 4π sphere of emission directions, we obtain a closed expression for \mathcal{I}_i :

$$\mathcal{I}_i(\mathbf{r}) = [\exp(ikr_i)/r_i] 4\pi \sum_{LM} F_{LM} (i)^L j_L(k|\mathbf{r} - \mathbf{r}_i|) Y_{LM}(\widehat{\mathbf{r} - \mathbf{r}_i}) \quad , \quad (36)$$

where we made use of the orthonormality of the spherical harmonics. From Eq.(36), we see that the holographic image is in fact a sum of standing partial waves. Notice that only the spherically symmetric ($L=0$) contribution in Eq.(36) actually peaks at the position of the scattering atom. For nonzero values of L , the j_L Bessel functions reach their maximum value away from the origin [18]. It is of some interest to note that in the region $k|\mathbf{r} - \mathbf{r}_i| \gg 1$, using the asymptotic expansion of the Bessel functions, $j_l(x) \sim \sin(x - l\pi/2)/x$, and with $(-1)^L Y_{LM}(\hat{\mathbf{r}}) = Y_{LM}(-\hat{\mathbf{r}})$, the image \mathcal{I}_i is a simple superposition of incoming and outgoing waves:

$$\begin{aligned} \mathcal{I}_i(\mathbf{r}) &\sim \frac{2\pi}{ik} \sum_{LM} F_{LM} \{ Y_{LM}(\widehat{\mathbf{r} - \mathbf{r}_i}) [\exp(ik|\mathbf{r} - \mathbf{r}_i|)/|\mathbf{r} - \mathbf{r}_i|] \\ &\quad - Y_{LM}(-\widehat{\mathbf{r} - \mathbf{r}_i}) [\exp(-ik|\mathbf{r} - \mathbf{r}_i|)/|\mathbf{r} - \mathbf{r}_i|] \} \\ &= \frac{2\pi}{ik} \{ f^{(eff)}(\widehat{\mathbf{r} - \mathbf{r}_i}; \hat{\mathbf{r}}_i) [\exp(ik|\mathbf{r} - \mathbf{r}_i|)/|\mathbf{r} - \mathbf{r}_i|] \\ &\quad - f^{(eff)}(-\widehat{\mathbf{r} - \mathbf{r}_i}; -\hat{\mathbf{r}}_i) [\exp(-ik|\mathbf{r} - \mathbf{r}_i|)/|\mathbf{r} - \mathbf{r}_i|] \} \quad . \end{aligned} \quad (37)$$

Returning to the problem of determining the spins, we assume that the scattering amplitudes f_s in Eq.(34) can be calculated numerically and can be used in the analysis of the hologram. By analyzing the holographic image of the spin holograms and by determining the particular mixture of partial waves in the image (s,p,d etc...), one can then, in principle, find the spin vectors.

However, regarding the use of calculated scattering amplitudes, we should remark that most numerical schemes assume spherical symmetry for the scatterer. While this assumption is mostly justified for the spin independent scattering amplitude f_0 , it is usually not for the spin dependent exchange scattering from the valence shell. As a matter of fact, the exchange scattering amplitude f_s is a quantity that is interesting in its own right, as we discuss in the

next section. Note that the projection holograms give the possibility of determining $f_s(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i)$ (and not just $|f_s|^2$). In many cases of interest, the spins are ordered in an arrangement with known projection onto a particular (α, j) -spin mode, or it might be possible to order the spins into such an arrangement, for example by applying an external magnetic field (in the case of C_{4v} -symmetry discussed above, one might be able to align the spins along the z-axis which is an A_2 -mode, or along the x-axis, which is an $(E, 1)$ -mode. Under those circumstances the spin directions of the (α, j) -mode are known and only the exchange scattering amplitude $f_s(\hat{\mathbf{R}}, \hat{\mathbf{r}}_i)$ needs to be determined in the expression for the projection hologram, which can be achieved by means of a least-square fit.

In fact, although holographic spin images are of interest, we think that the most efficient and accurate spin determinations will involve fitting the hologram in a scheme that is similar to the procedure to determine f_s , with the coefficients of the different spin modes of the same (α, j) -representation as additional parameters to be determined. One can devise an iterative scheme to determine f_s and the spins: Use the calculated f_s to estimate the atomic spin vectors. Then, use the obtained spins to refine the scattering amplitude f_s through a least-square fit. The resulting f_s is then used to obtain a better fit of the spins, and so on, until convergence is reached.

VI. NOTE ON RARE EARTHS AND SPIN HOLOGRAPHY

In our discussion in the preceeding sections we assumed that the spin dependent part of the coherent scattering amplitude, see Eq. (5), for an atom ‘j’ was of the form $F_s \sim \sigma \cdot \langle \mathbf{S}_j \rangle f_s(\mathbf{k}, \mathbf{k}')$. This is a good approximation, however, only when we can assume that the spin density of atom j is of the form $\mathbf{s}_j(\mathbf{r}) = \mathbf{S}_j \rho_j(\mathbf{r})$; that is, that the orbital angular moments are ‘quenched’. This is usually a good assumption for transition elements in crystals; but not for rare earth (or actinide) elements where there are strong spin-orbit correlations. In that case, we obtain (see appendix)

$$F_s(\mathbf{k}, \mathbf{k}') = \sigma \cdot \sum_{K=0, M=-K}^{2l, K} A_K T_{KM}^*(\hat{\mathbf{k}}, \hat{\mathbf{k}}') \langle \mathbf{S}_j \mathcal{Y}_{KM}(\mathbf{L}_j) \rangle . \quad (38)$$

In the equation F_s is the spin-dependent coherent exchange contribution to the scattering amplitude ($\mathbf{k} \rightarrow \mathbf{k}'$) from a rare earth ion 'j' where Russel-Saunders coupling is assumed to apply with \mathbf{S}_j and \mathbf{L}_j the good spin and orbital angular momentum of the ion. $\mathcal{Y}_{KM}(\mathbf{L})$ is an irreducible tensor constructed from the angular momentum operator \mathbf{L} , and $T_{KM}(\hat{\mathbf{k}}, \hat{\mathbf{k}}')$ is an irreducible tensor depending on $\hat{\mathbf{k}}$ and $\hat{\mathbf{k}}'$. A_K involves radial exchange integrals as well as other coupling coefficients (see appendix).

For the rare earths $l=3$ and Eq.(38) gives F_s in terms of the scattering from the six multipole moment tensors of the uncompensated spin distributions of the rare earth ion.

If the spin and orbits are not coupled then

$$F_s = \sigma \cdot \langle \mathbf{S}_j \rangle \sum_{K, M} A_K T_{KM}^*(\hat{\mathbf{k}}, \hat{\mathbf{k}}') \langle \mathcal{Y}_{KM}(\mathbf{L}_j) \rangle , \quad (39)$$

and we have the quenched orbital result of Eq.(5).

In Eqs.(38) and (39), $K = 0$ is the isotopic scattering term, $T_0(\hat{\mathbf{k}}, \hat{\mathbf{k}}') = f_0(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}')$, T_1 the vector term, $T_1(\hat{\mathbf{k}}, \hat{\mathbf{k}}') = \hat{\mathbf{k}} a_{11}(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') + \hat{\mathbf{k}}' a_{12}(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') + \hat{\mathbf{k}} \times \hat{\mathbf{k}}' a_{13}(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}')$. Similarly, $T_{20}(\hat{\mathbf{k}}, \hat{\mathbf{k}}') = (\hat{\mathbf{k}}_z^2 - 1/3) a_{21}(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') + (\hat{\mathbf{k}}_z'^2 - 1/3) a_{22}(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') + (\hat{\mathbf{k}}_z \hat{\mathbf{k}}_z' - 1/3 \hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') a_{23}(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}'), \dots$

In the hypothetical case of no spin-orbit coupling then the spin $\langle \mathbf{S}_j \rangle$ would be free to vary independent of the anisotropic charge distribution and would correspond to our previously discussed theory. In fact, however, magnetic studies show [20] that $J = |\mathbf{L} + \mathbf{S}|$ retains its Hunds rule ground state value, $J = L + S$, for the second half shell, and $J = L - S$ for the first half shell in the crystal (except Eu^{3+} and Ce^{3+}) to a good approximation.

In that case Eq.(38) becomes

$$F_s = \sum_{K'=K-1, K=0, M'}^{K+1, 2l} A_K B_{KK'} (\sigma \otimes T_K(\hat{\mathbf{k}}, \hat{\mathbf{k}}'))_{K'M'}^* \langle \mathcal{Y}_{K'M'}(\mathbf{J}) \rangle , \quad (40)$$

where $B_{KK'}$ is another coupling coefficient (see Appendix) and

$$(\sigma \otimes T_K(\hat{\mathbf{k}}, \hat{\mathbf{k}}'))_{K'M'} \equiv \sum_{\mu, \nu} C_{\mu \nu}^{1 K K'} \sigma_{\mu} T_{K\nu}(\hat{\mathbf{k}}, \hat{\mathbf{k}}') . \quad (41)$$

Discussion. Now, instead of just $\langle \mathbf{S} \rangle$ or $\langle \mathbf{J} \rangle$ to represent the spin dependent coherent exchange scattering, there are seven, $2l+1$, multipole moment distributions $\langle \mathcal{Y}_{KM}(\mathbf{J}) \rangle$ giving $(2l+1)^2 = 49$ parameters determining the scattering. Gd^{3+} is uniquely simple because it is spherically symmetric so that in Eq.(38) $\langle \mathcal{Y}_{KM}(\mathbf{L}) \rangle = 7\delta_{K0}$ and thus $F_s \sim \sigma \cdot \langle \mathbf{J} \rangle f_0(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}')$. For the other rare earths however, and for $E_{k,k'} \simeq 100$ eV, tensor components up to $K \simeq 3, 4$ make important contributions to F_s .

The question now arises, are there good reasons to measure and analyze ‘spin holograms’ of rare earth ions, given their evident complexity? We think there is because of the relation of such scattering measurements to the RKKY mechanism responsible for the rare earth ion interactions in solids. The interaction arises from the exchange scattering of conduction electrons with 4f shell electrons of an ion inducing a stationary spin polarization wave in the surrounding conduction band. The role of the anisotropic, higher moment terms, Eq.(40), in F_s on the RKKY-interaction have not been investigated as thoroughly as their importance warrants. Frederick Specht [21] in an early investigation retained only terms up to $K = 2$ and found quite large effects. Thus for two nearest neighbor *Tb* ions whose moments were aligned along the inter-ionic direction he obtained $E(0) = -6.6k_B$, while if the moments are perpendicular to that direction he obtained $E(\pi/2) = -8.1k_B$, a 20% variation; while for *Tm* ions the numbers were $E(0) = -1.2k_B$, $E(\pi/2) = -0.7k_B$, a 50% variation. (Note that for the cigar shaped spin distribution of *Tb* $|E(\pi/2)| > |E(0)|$, while the opposite held for the pancake shaped *Tm* spin distributions).

We think that spin holographic experiments on a ferromagnetic surface layer of terbium, for example, as a function of temperature, magnetic field, and k of the photoelectron, revealing the effects of the moments of various orders, would be very interesting.

Measurement Theory. An interesting question is what measurement must be made to determine the quantum state of a system [22]. Consider a system with angular momentum J . In general its state is determined by its density matrix $\rho_{M,M'}$. What measurements can be made to determine the $(2J+1)^2 - 1 = 4J(J+1)$ real numbers required to specify ρ ? In reference [22] we outlined one method using ‘Feynman filters’; but we also men-

tioned that Fano [23] had shown that measurement of the $4J(J+1)$ expected multipole moments $\langle \mathcal{Y}_{KM}(\mathbf{J}) \rangle$, $K = 1, \dots, 2J$ also would suffice. In our work here we have seen that the $\langle \mathcal{Y}_{KM}(\mathbf{J}) \rangle$'s could be determined (in principle) for $K \leq 2l+1$, or $K \leq 2L+1$ by means of coherent electron scattering, the higher order multipoles vanish. Thus for Gd only the first order moment, $\langle \mathbf{J} \rangle$ could be so determined, while for the other rare earths much more state information can be determined, but not enough to fully determine the state.

VII. CONCLUSIONS

The main message of this paper is that we can construct holograms of the atomic spins in the near-neighborhood of a photoelectron emitting atom from the angularly resolved photoelectron emission intensities. For the important case of photoelectrons emitted by source atoms with C_{nv} -environment, we show how these spin holograms can be constructed using symmetry. Furthermore, we discuss schemes to holographically image the spins and to extract accurate spin information from the spin holograms.

The photons used to emit the electrons that give the intensity holograms for the purpose of spin holography can be, *but do not have to be*, circularly polarized. Incident linearly polarized, or indeed even *unpolarized* light would also serve the purpose. This statement could appear somewhat puzzling because it seems to imply that it is unnecessary to polarize the primary photoelectron waves in order to probe the spins of the nearby scattering atoms. We show that the surprising ability of unpolarized light to record spin information in the photoelectron intensity is a consequence of the spin-orbit correlations in the initial inner-core electron states from which the photoelectron is emitted. The spin-orbit correlations give a finite interference contribution to the spin of electrons emitted in different directions. It is the interference of the photoelectron emitted in the $\hat{\mathbf{R}}$ -direction, and that emitted in the direction of atom i and then scattered into the $\hat{\mathbf{R}}$ -direction, that gives the holographic fringes in the photoelectron intensity. By virtue of exchange scattering the holographic fringes are then sensitive to the spin of the scattering atom, with contributions proportional to the

scalar product of the atomic spin and the interference contribution to the photoelectron spin, $\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{r}}_i\rangle\rangle$. Note that it is not, as one might have surmised, the spin of the photoelectron wave emitted in the \mathbf{r}_i -direction, $\langle\langle\hat{\mathbf{r}}_i|\sigma|\hat{\mathbf{r}}_i\rangle\rangle$, that is of importance. Indeed, under some conditions, this primary wave spin polarization can vanish, implying unpolarized electrons impinging on the scattering atom, while $\langle\langle\hat{\mathbf{R}}|\sigma|\hat{\mathbf{r}}_i\rangle\rangle \neq 0$, implying a spin dependent photoelectron intensity hologram. In the paper, we calculate the spin interference matrix element for photoemission from a $p_{1/2}$ shell, by absorbing photons of arbitrary polarization.

Furthermore, to obtain an actual spin hologram from the photoelectron intensity, it is necessary to ‘separate out’ the spin dependent holographic fringes, which typically contribute 5 % or less, from the rest of the hologram. In this paper, we point out how this separation can be obtained using symmetry if the source atom is in a site of C_{nv} -symmetry. Specifically, we show that a group theoretical projection of the emission intensity onto the irreducible (α, j) -representation gives a hologram of the projection of the spin arrangement of the nearby atoms onto the (α, j) -spin mode. This way, one can obtain almost all components of the spin vectors (all but one for the n -atom star and all but three for the $2n$ -atom star, as explained in the text).

The advantage of measuring emission intensities, rather than having to spin analyze the emitted electrons, is considerable: one does not lose the usual factor of 10^4 in intensity due to the low efficiency of the electron polarimeters. Consequently, we expect that the proposed experiments are quite feasible at a synchrotron source and it is possible that the data have already been obtained and only need to be analyzed.

To conclude, we repeat that spin holography is a surface probe which probes the average short-range magnetic environment of the photoelectron source atom. By using atoms adsorbed on a surface as sources, spin holography can reveal the influence of these adsorbates on the spins of the neighboring substrate atoms. It can then provide detailed information on many interesting systems, such as adsorbed oxygen atoms quenching the magnetism of a nickel surface, or iron atoms inducing moments on neighboring palladium substrate atoms (superparamagnetism). Alternatively, substrate atoms can be used as sources to image the

spins of objects adsorbed on the surface, such as small molecules. Finally, spin holography should be useful for surface studies of magnetic systems with large and complicated unit cells of N spins, such as garnets. (Spin holography gives information about the N spins as a function of the N positions, whereas the usual diffraction techniques give the N^2 spin-spin correlation as functions of the N^2 relative positions).

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APPENDIX

We shall sketch the derivation of Eqs.(38-40).

If \mathbf{s}_c is the spin of the positive energy ‘free’ electron and \mathbf{s}_i that of a bound electron then the spin exchange operator is $1/2 + 2\mathbf{s}_c \cdot \mathbf{s}_i$, and the exchange scattering amplitude for free electron momentum \mathbf{k} and bound electron in orbital $|u_{lm}\rangle$ going to a free electron momentum \mathbf{k}' and bound electron $\rightarrow |u_{lm'}\rangle$ is

$$f_s = (1/2 + 2\mathbf{s}_c \cdot \mathbf{s}_i) \mathcal{S}(\mathbf{k}, u_{lm}; \mathbf{k}', u_{lm'}) , \quad (42)$$

where

$$\mathcal{S} = - \int \exp[-i\mathbf{k}' \cdot \mathbf{r}_2] u_{lm'}^*(\mathbf{r}_1) \frac{e^2}{r_{12}} u_{lm}(\mathbf{r}_2) \exp[i\mathbf{k} \cdot \mathbf{r}_1] d^3r_1 d^3r_2 , \quad (43)$$

is the exchange integral.

Expanding the integral in partial waves we get

$$\mathcal{S} = \sum_{p, p', \lambda} E(l, p, k; l, p', k'; \lambda) \sum_{q, q', \mu} \langle l, m' | Y_{pq} | \lambda \mu \rangle \langle \lambda \mu | Y_{p'q'} | lm \rangle Y_{p'q'}^*(\hat{\mathbf{k}}') Y_{pq}^*(\hat{\mathbf{k}}) , \quad (44)$$

where E involves radial overlap integrals

$$E(l, p, k; l, p', k'; \lambda) = - (i)^{p-p'} \frac{(4\pi)^3 e^2}{(2p+1)(2p'+1)(2\lambda+1)} \int j_{p'}(k' r_2) g_l(r_1) \frac{r_1^\lambda}{r_1^{\lambda+1}} g_l(r_2) j_p(k r_1) r_1^2 r_2^2 dr_1 dr_2 , \quad (45)$$

with j_p the spherical bessel function and $u_{lm}(\mathbf{r}) = g_l(r) Y_{lm}(\hat{\mathbf{r}})$.

Now making use of the Racah-Wigner techniques ([24]) we obtain

$$\begin{aligned} \mathcal{S} = & \sum_{p, p', \lambda} \langle l || Y_p || \lambda \rangle \langle \lambda || Y_{p'} || l \rangle \sqrt{2\lambda+1} E(l, p, k; l, p', k'; \lambda) \\ & \cdot \sum_{KM} (-)^K \sqrt{2K+1} \left\{ \begin{matrix} l & p & \lambda \\ p' & l & K \end{matrix} \right\} \left[\left(Y_{p'}(\hat{k}') \otimes Y_p(\hat{k}) \right)_M^K \right]^* C_m^l K M m' . \end{aligned} \quad (46)$$

We now replace the vector addition coefficient in (46) in accordance with $C_m^l K M m' = \langle lm' | \mathcal{Y}_{KM}(\mathbf{l}_i) | lm \rangle / \langle l || \mathcal{Y}_K || l \rangle$, where $\mathcal{Y}_{KM}(\mathbf{l})$ is an irreducible tensor of rank K constructed from the angular momentum operators \mathbf{l} .

The exchange scattering operator is now obtained by multiplying \mathcal{S} by the spin exchange operator and summing over the n electrons in the $4f$ shell:

$$\begin{aligned} F_x &= \sum_i^n f_s(i) \\ &= \sum_{K=0, M=-K}^{2l, K} A_K T_{KM}^*(\hat{\mathbf{k}}, \hat{\mathbf{k}}') \langle (1/2 + 2\mathbf{s}_c \cdot \frac{\mathbf{S}}{\nu}) \mathcal{Y}_{KM}(\mathbf{L}) \rangle, \end{aligned} \quad (47)$$

where \mathbf{L} , \mathbf{S} are the operators for the total orbital angular momentum and spin of the $4f$ electrons (we assume Russel-Saunders coupling with good L and good S). Also, we have supposed the Russel-Saunders ground state values of L and S and we made the substitution

$$\begin{aligned} &\sum_{i=1}^n (1/2 + 2\mathbf{s}_c \cdot \mathbf{s}_i) \mathcal{Y}_{KM}(\mathbf{l}_i) \\ &= \pm \frac{\langle lL || \sum_{i=1}^\nu \mathcal{Y}_K(\mathbf{l}_i) || lL \rangle}{\langle L || \mathcal{Y}_K(\mathbf{L}) || L \rangle} \langle (1/2 + 2\mathbf{s}_c \cdot \frac{\mathbf{S}}{\nu}) \mathcal{Y}_{KM}(\mathbf{L}) \rangle, \end{aligned} \quad (48)$$

where $\nu = n$ for less than half filled shell, $\nu = N - n$ for more than half filled. For more than half filled shell we must choose the minus sign in (47) and then add $\frac{N}{2}\delta_{K0}$ to the spin independent term (i.e. rather than $-(\frac{\nu}{2} + 2\mathbf{s}_c \cdot \mathbf{S})$ for the $K = 0$ term we get $(\frac{N-\nu}{2} - 2\mathbf{s}_c \cdot \mathbf{S})$ for that term). Thus finally

$$\begin{aligned} F_x &= \eta \sum_{p, p', \lambda} E(l, p, k; l, p', k'; \lambda) \langle l || Y_p || \lambda \rangle \langle \lambda || Y_{p'} || l \rangle \sqrt{2\lambda + 1} \\ &\quad \sum_{KM} (-)^K \sqrt{2K + 1} \left\{ \begin{matrix} l & p & \lambda \\ p' & l & K \end{matrix} \right\} [Y_{p'}(\hat{\mathbf{k}}') \otimes Y_p(\hat{\mathbf{k}})]_{KM}^* \\ &\quad \frac{\langle lL || \sum_{i=1}^\nu \mathcal{Y}_K(\mathbf{l}_i) || lL \rangle}{\langle L || \mathcal{Y}_K || L \rangle} (1/2 + 2\mathbf{s}_c \cdot \frac{\mathbf{S}}{\nu}) \mathcal{Y}_{KM}(\mathbf{L}), \end{aligned} \quad (49)$$

where η is (+/-) for (less than/more than) half filled shell systems. The meaning of $A_K T_{KM}^*(\hat{\mathbf{k}}, \hat{\mathbf{k}}')$ in Eq.(47) can then be obtained by compairing with Eq.(49).

Finally we suppose that J is also a good quantum number (with $J = L - S$ for the first half and $J = L + S$ for the second half rare earths). Then within the manifold of good L, S and J

$$\mathcal{Y}_{KM}(\mathbf{L}) = (-)^{L+S+J+K} \sqrt{(2L+1)(2J+1)} \left\{ \begin{matrix} J & S & L \\ L & K & J \end{matrix} \right\} \frac{\langle L || \mathcal{Y}_K(\mathbf{L}) || L \rangle}{\langle J || \mathcal{Y}_K(\mathbf{J}) || J \rangle} \mathcal{Y}_{KM}(\mathbf{J}), \quad (50)$$

and this may be substituted into the spin independent term in Eq.(47). Similarly,

$$\begin{aligned}
& \sum_M T_{KM}^*(\hat{\mathbf{k}}, \hat{\mathbf{k}}') \sigma \cdot \mathbf{S} \mathcal{Y}_{KM}(\mathbf{L}) \\
&= \sum_{K'\nu} \left[\sigma^{(1)} \otimes T_K \right]_{\nu}^{K'*} \cdot \left[\mathbf{S}^{(1)} \otimes \mathcal{Y}_K(\mathbf{L}) \right]_{\nu}^{K'} \\
&= \sum_{K'\nu} \left[\sigma^{(1)} \otimes T_K(\hat{\mathbf{k}}, \hat{\mathbf{k}}') \right]_{\nu}^{K'} \begin{bmatrix} S & L & J \\ 1 & K & K' \\ S & L & J \end{bmatrix} \langle S || S || S \rangle \langle L || \mathcal{Y}_K(\mathbf{L}) || L \rangle \frac{\mathcal{Y}_{K'\nu}(\mathbf{J})}{\langle J || \mathcal{Y}_{K'}(\mathbf{J}) || J \rangle} , \quad (51)
\end{aligned}$$

and substituting (51) into Eq.(47) gives Eq.(40) of the text.

TABLES

TABLE I. Character table of the group C_{4v}

C_{4v}	E	$2C_4$	C_2	$2\sigma_v$	$2\sigma_d$
A_1	1	1	1	1	1
A_2	1	1	1	-1	-1
B_1	1	-1	1	1	-1
B_2	1	-1	1	-1	1
E	2	0	-2	0	0

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images, one can determine the partial wave mixture by holographically transforming with anisotropy-correcting kernels, finding the kernel that gives the most s-wave like image. Alternatively, one can try to ‘isolate’ a single component of the spin vector of atom i , by using a kernel in the holographic transform that gives a peak at \mathbf{r}_i for one spin direction, but vanishes at $\mathbf{r} = \mathbf{r}_i$ for the spin component in the perpendicular direction, see also ref. [8].

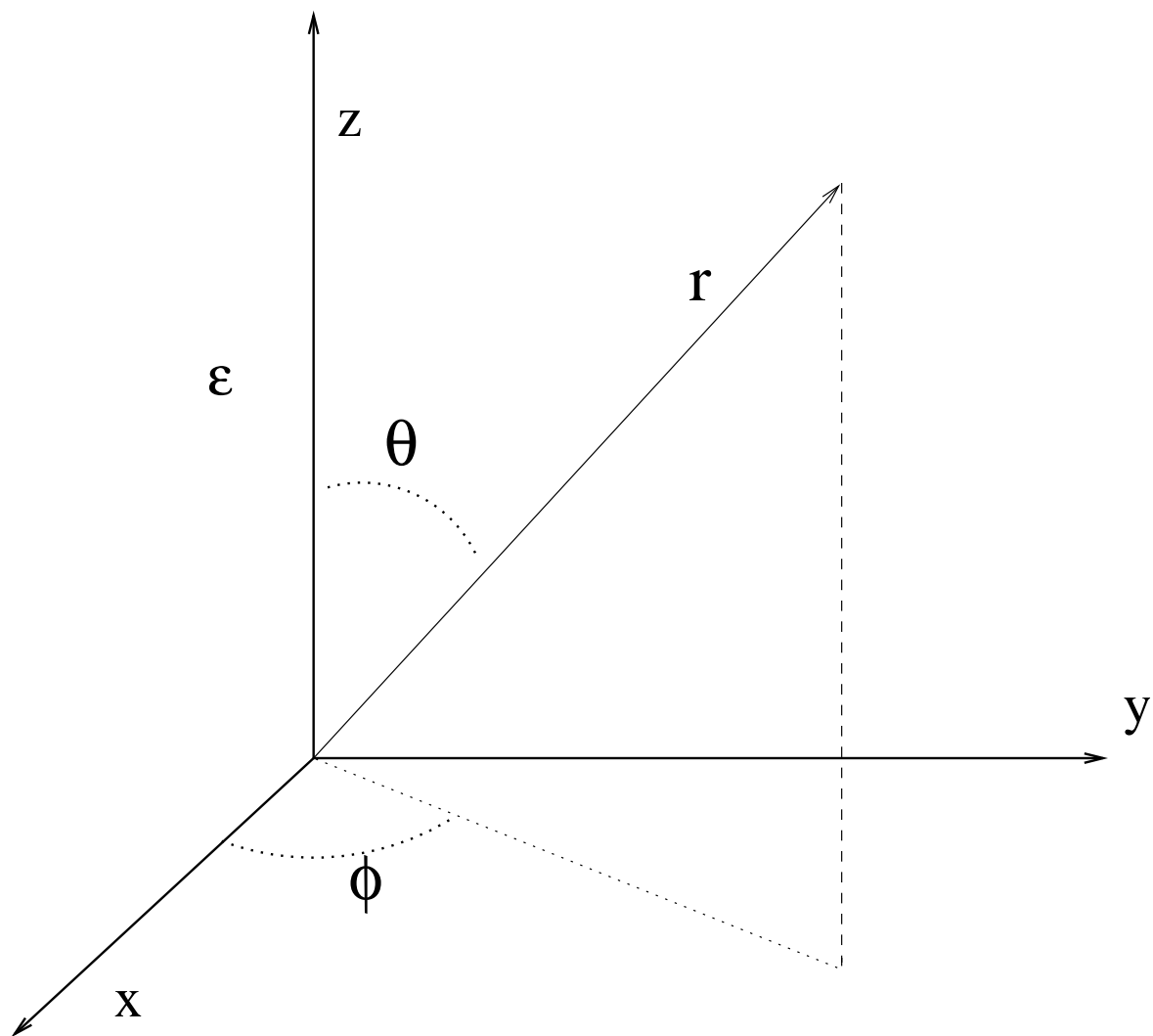
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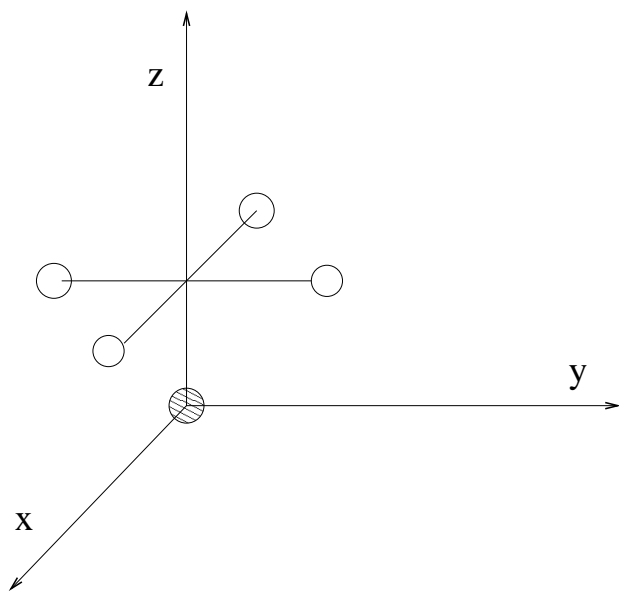
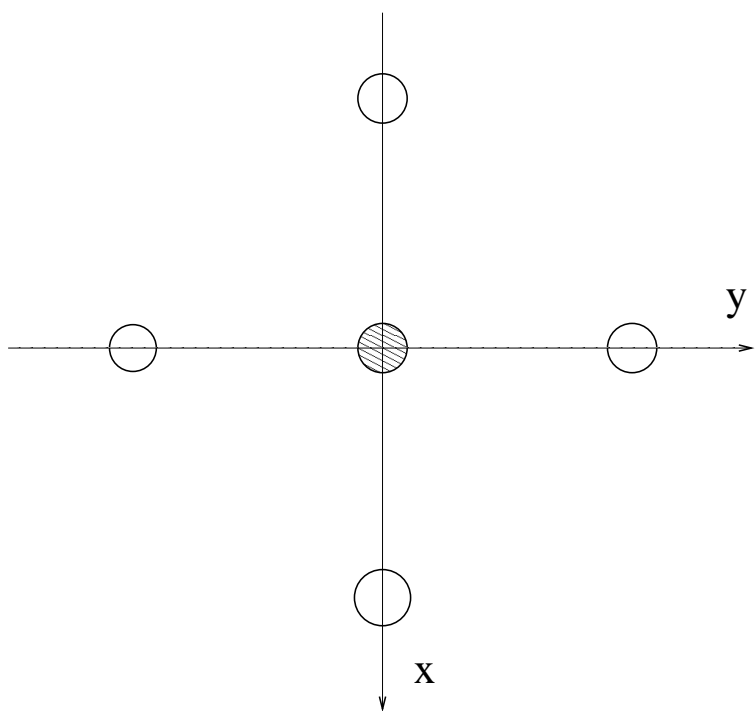
FIGURE CAPTIONS

Figure 1: Reference frame used in calculating the spinors of the electrons emitted from a $p_{1/2}$ -shell (section II). The photon polarization vector, $\hat{\epsilon}$, is parallel to the $\hat{\mathbf{z}}$ -direction.

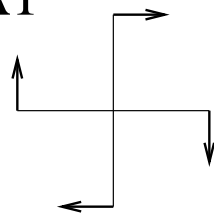
Figure 2: 4-atom environment of electron emitting atom (denoted by the shaded sphere). The symmetry-group of the environment is C_{4v} , with the C_4 -axis ($= \hat{\mathbf{z}}$ -axis) perpendicular to the plane of the four atoms.

Figure 3: The group theoretical modes of the spin vectors of the C_{4v} -environment shown in Fig.2. The spins are shown in the xy-plane, \odot indicates an up-spin (positive $\hat{\mathbf{z}}$ -direction), and \otimes indicates a ‘down’-spin (negative $\hat{\mathbf{z}}$ -direction).

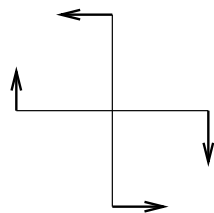




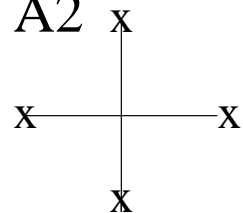
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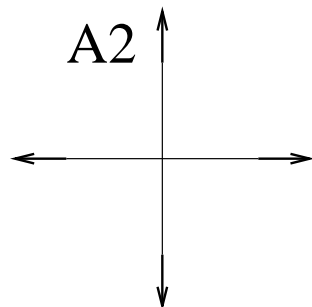
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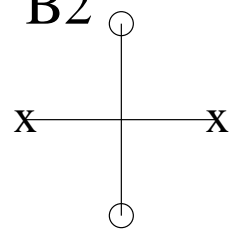
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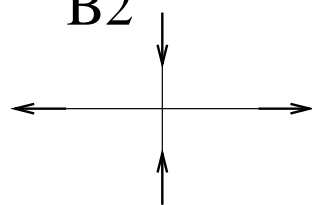
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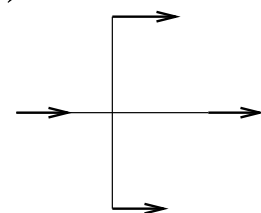
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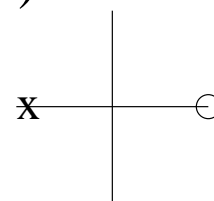
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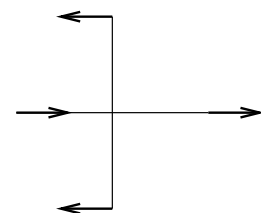
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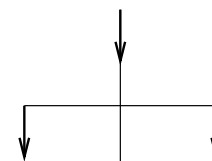
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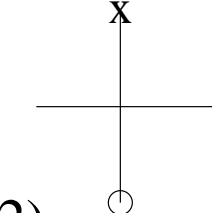
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(E,2)



(E,2)



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